Docket No.: 256241US0PCT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:
Naoto OHTA, et al.

SERIAL NO: 10/501,333

GROUP: 4191

CONFIRMATION NO. 9835

EXAMINER: Best, Z.

FILED: July 23, 2004

FOR: ANODE MATERIAL FOR LITHIUM ION SECONDARY BATTERY

DECLARATION UNDER 37 C.F.R. § 1.132

COMMISSIONER FOR PATENTS
ALEXANDRIA, VIRGINIA 22313

Sir:

Now comes Naoto OHTA who deposes and states that:

1. I am a graduate of Toyohashi University of Technology and received my Master's degree in the year 1989.

2. I have been employed by Toyo Tanso Co. LTD for 13 years and currently serve as a Chief in the field of fundamental carbon-technology.

3. I am a named inventor in the present application.

4. The following experiments were carried out by me or under my direct supervision and control.

5. Coated graphite powders of the present invention are made by dry-blending, where, *e.g.*, 100 parts by weight of natural graphite powder having an average interlayer spacing d(002) of 0.3354 (defined by the Gakushin-method) was dry-blended with polyvinyl alcohol (PVA) powder. The mixed powders were then heat treated. Table 1 shows the average interlayer spacing d(002) of graphite powder coated with the polyvinyl alcohol as a function of the amount of polyvinyl alcohol added:

Table 1: Average interlayer spacing d(002) of anode material before and after resin coating.

Added amount of PVA (parts by weight with respect to 100 parts by weight of natural graphite)	d(002) (nm)
0 (no coating)	0.3354
10	0.3354
50	0.3355
100	0.3354

*Although 50 parts by weight of PVA is adopted in the examples of the present application, it is understood that d(002) does not change even in the case of 100 parts by weight.

As shown in Table 1, the d(002) of the graphite powder before coating is substantially identical with d(002) after coating (*i.e.* changes less than 0.0005 nm).

6. Graphite powder was also prepared by the process of Example 1 disclosed in U.S. Patent 6,596,437. The average interlayer spacing d(002) of the graphite powder without a resin coating was determined by X-ray diffraction: d(002) = 3.35 Å. The average interlayer spacing d(002) was then determined for the graphite powder coated with a resin: d(002) = 3.41 Å. Thus, a substantial increase in the interlayer spacing of the graphite powder prepared by the process of Example 1 in U.S. Patent 6,596,437 is observed.

7. X-ray diffraction can detect the values for interlayer spacing of crystalline materials (*e.g.* graphite), which knowledgeable practitioners in the field recognize. This analytical technique is therefore able to measure changes in interlayer spacing of a crystal lattice upon perturbation of the crystal lattice from an initial state. If the interlayer spacing increases for a crystalline substrate upon a perturbation, then size of the particles occupying the crystal lattices is likely increasing.

8. When applied to graphite, the interlayer spacing is first measured in graphite before treatment, *e.g.* coating with a thermoplastic resin. A second measurement of the interlayer spacing is taken upon treating the graphite powders with a thermoplastic resin. If the value of the interlayer spacing increases, then the thermoplastic resin coats the outer surface of the graphite powder, thereby causing the diameter of the graphite particle to increase. On the other hand, if no difference is measured between the values for the interlayer spacing upon coating

the particles, then the coating is not forming a shell around the outer surface of the graphite particles.

9. In the case of the dry-blending of the present invention, the results show that the average interlayer spacing of the graphite powder made by dry-blending does not change (*i.e.* at most 0.0005 nm) when resin is applied to the graphite powder. Thus, dry-blending as in the present invention does not result in forming an outer shell of thermoplastic resin around the outer surface of the graphite particles. However, evidence that the thermoplastic resin is present within the fine pores in the graphite particles is seen in the irreversible capacities of batteries where the graphite particles are used as anodes.

10. For the battery of Example 1 of the present invention, the graphite particles used in the anodes are coated with a thermoplastic resin. The irreversible capacity for this battery is 28.2 mAh/g. On the other hand, the battery of Comparative Example 7 was made in the same way as the battery of Example 1 except that the graphite was not coated. The irreversible capacity for this battery was 78.2 mAh/g. Thus, a significant decrease is observed in the irreversible capacity when a coated graphite is used. As noted in the present application, decomposition of the electrolyte results in a large irreversible capacity. Taken together, the reduced irreversible capacity shows that that the thermoplastic resin coating is present in the graphite powder, while the X-ray diffraction data show that the thermoplastic resin coats the fine pores present in the graphite powder rather than forming a shell around the graphite powder.

11. On the other hand, the graphite powder made by the process of Example 1 in U.S. Patent 6,596,437 exhibits an increased average interlayer spacing. As noted previously, the average interlayer spacing d(002) of the graphite powder without a resin coating was 3.35 Å, and the average interlayer spacing d(002) was then determined for the graphite powder coated with a resin was 3.41 Å. This corresponds to an increase of 0.006 nm. The Abstract of

U.S. Patent 6,596,437 states: "The active material includes a crystalline carbon core; and an amorphous or turbostratic carbon shell evenly covering the crystalline carbon core, the carbon shell..." Accordingly, the graphite powders of U.S. Patent 6,596,437 have an even shell of resin on the outer surface of the graphite particles, rather than only filling any fine pores present within the graphite.

12. The undersigned petitioner declares further that all statements made herein of his own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

13. Further deponent saith not.

Signature Naoto Ohta

Date 2009. 11. 7

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